

# Grain boundary roughening transitions

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## Abstract

We consider the roughening of small angle grain boundaries consisting of arrays of dislocations and found two transitions, corresponding to fluctuations of the dislocations along and perpendicular to the boundaries. The latter contributes to a large scale fluctuation of the orientation of the crystal but the former does not. The transition temperatures of these transitions are very different, with the latter occurring at a much higher temperature. Order of magnitude estimates of these temperatures are consistent with recent experimental results from elasticity and X-ray measurements in solid  $^4\text{He}$ .

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Since the discovery of an increase in torsional oscillator frequency in solid  $^4\text{He}$  at around 200 mK[1], there have been renewed interests in its low temperature physical properties. Much recent focus is on the role played by defects in this system.[2, 3, 4, 5, 6, 7, 8] For example, it is suggested that large angle grain boundaries can exhibit superfluid behavior[4, 8]. X-ray measurements[2] found a change in the orientational fluctuation of the crystallites at around 1.75 K. Recently Day and Beamish[3] found a change in the shear modulus with the same temperature dependence as that for the decoupling in torsional oscillators. They ascribe this to a change in the mobility of dislocations.

There usually is a **finite** density of dislocations and it is important to consider the long range elastic interaction between them. The simplest arrangement of a collection of dislocations comes from the small angle grain boundary (GB). These boundaries are pinned by the Peierls potential even in the absence of additional impurities. There has been much interest in the study of the pinning of an elastic two dimensional interface. Above the roughening transition temperature  $T_R$  the free energy to create a step becomes zero and the interface is depinned. This roughening transition temperature is a function of the strength of the pinning potential. Even as the strength of the pinning potential approaches zero,  $T_R$  remains **finite**.

In this paper we examine if a GB can roughen and found that for a "electrically neutral" system, there are **two** roughening transitions, corresponding to the motion of the dislocations parallel and perpendicular to the boundary. The latter contributes to a large scale fluctuation of the orientation of the crystal as is observed in the X-ray experiments[2]. The other transition occurs at a much lower temperature, does not contribute to the large scale angular fluctuations but, because of the change in mobility of the dislocation, can cause a change in the elastic coefficients, similar to that observed experimentally[3]. Order of magnitude estimates of these temperatures are consistent with experimental results from elasticity[3] and X-ray measurements[2] in solid  $^4\text{He}$ . We now describe our results in detail.

A small angle GB consists of an array of dislocations with parallel Burger's vectors  $\mathbf{b}$ . The trajectory of a dislocation can be represented by the positions of elements separated by lattice constants  $a_z = a_0$  along it. We describe the configuration of the GB by the positions  $\mathbf{c}_j$  of the elements of the dislocations. The elastic interaction between two elements of the

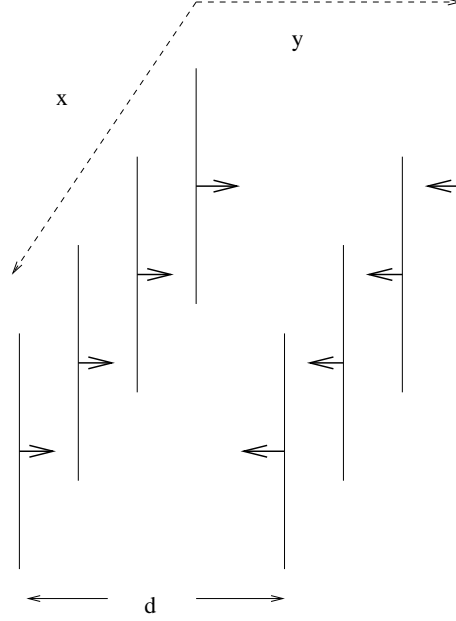


FIG. 1: Schematic diagram of the grain boundaries.

dislocations is given by the formula

$$V = \kappa/(4\pi)[\mathbf{b} \cdot \mathbf{b}'/R + \mathbf{b} \cdot \mathbf{R}\mathbf{b}' \cdot \mathbf{R}/R^3] \quad (1)$$

where  $\mathbf{R}$  is the distance between the elements. For simplicity we assume an elastically isotropic system. In terms of the Lamé constants  $\lambda$  and  $\mu$ ,  $\kappa = 4a_0^2\mu(\mu + \lambda)/(2\mu + \lambda)$ . In addition, for each dislocation there is a core energy contribution  $V^c = (E_c/a_0)L$  that is proportional to its length  $L = \int dl$ , where  $dl = (dc_x^2 + dc_y^2 + dc_z^2)^{1/2}$ . Finally there is the pinning energy which we assume to be of a Peierl's form  $\sum_j U_x \cos(c_{xj}/a_0) + U_y \cos(c_{yj}/a_0)$ .

We take the dislocation line along  $z$ ; the Burger's vectors along  $y$ . Thus the GB is in the  $xz$  plane. This is illustrated in fig. 1. The roughening transition depends on the form of the energy change for small deviations  $\delta\mathbf{c}_j$  in the location of the dislocations. This deviations of the dislocations are along the  $x$  (parallel to the GB) and  $y$  (perpendicular to the GB) directions only. The fluctuation in the location of the dislocations  $\delta\mathbf{r}$  does not have a component along  $z$ . The energy change determines the "elastic" properties of the GB. We shall be interested in the Fourier transform  $\delta\mathbf{c}(\mathbf{q}) = \sum_j \delta\mathbf{c}_j \exp(-i\mathbf{q} \cdot \mathbf{c}_j)/\sqrt{N}$ . The GB is in the  $xz$  plane. Hence the wave vector  $\mathbf{q}$  is in the  $xz$  plane only. The distance between the dislocations  $a_x = a_0/\theta$  is related to the misorientation angle  $\theta$  between the grains. We consider the simplest "electrically neutral" system with zero total Burger's vectors consisting

of two grain boundaries of opposite Burger's vectors; one at  $y=0$  and the other one at  $y=d$ . We focus on the lowest energy acoustic mode so that the displacements on the two boundaries are the same.

A single grain boundary corresponds to two **infinite** crystals of different orientation joining together whereas a grain will be surrounded by boundaries of opposite orientations. We find that a **single** grain boundary does **not** roughen and thus focus on the pair, which also occur physically.

The contribution to the energy change from the core energy is equal to

$$\delta V^c = \sum_{\mathbf{q}} 0.5 E_c q_z^2 [|\delta c_x(\mathbf{q})|^2 + |\delta c_y(\mathbf{q})|^2]. \quad (2)$$

We next look at the contribution from  $V$ . From eq. (1) the change in  $V$  is given by

$$\delta V = \kappa / (4\pi) b^2 \sum_{i,j=x,y} \delta c_{iq} \delta c_{j,-q} (D_{ij}^s + D'_{ij}) \quad (3)$$

where the contribution from dislocations in the same GB (self) is

$$D_{ij}^s = \sum_R [1 - \cos(iq \cdot R)] [\nabla_i \nabla_j 1/R + \nabla_i \nabla_j y^2/R^3] \quad (4)$$

Similarly the contribution from the interaction energy between dislocations on different boundaries is given by

$$D'_{ij} = - \sum_R [1 - \cos(iq \cdot R)] [\nabla_i \nabla_j 1/R' + \nabla_i \nabla_j U] \quad (5)$$

$U = (y + d)^2/R'^3$ ,  $r' = (R^2 + d^2)^{1/2}$ .  $D$  is very similar to the dynamical matrix for the two dimensional Wigner crystal which has been considered in detail with the Ewald sum technique by Bonsall and Maraduddin[9]. The two dimensional sums  $D^s$ ,  $D'$  can be evaluated in the same manner.

We find in the long wavelength limit the x mode and the y mode are not coupled. For the "x" mode, the energy change is

$$\delta V^c + \delta V^x = (C_x q_x^2 + C_z q_z^2) |\delta c_x(\mathbf{q})|^2, \quad (6)$$

$C_x = \kappa b^2 (\pi^{-1/2} + 2) d / (a_x a_z)$ ,  $C_z = 0.5 E_c - 0.21 \kappa b^2 / (a_z \pi)$ . Without the core energy contribution  $C_z$  is less than zero and the lattice is unstable. This is consistent with the fact that

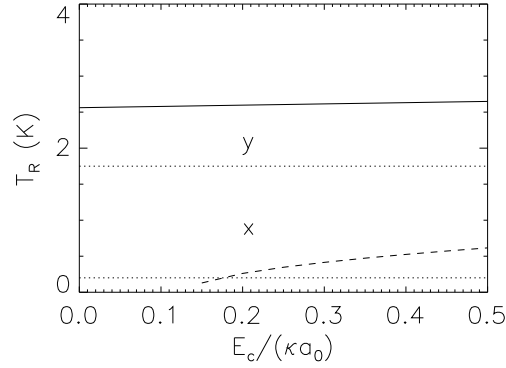


FIG. 2: The roughening temperature in units of Kelvin for the y mode (solid line) and the x mode (dashed line) as a function of the core energy normalized by  $\kappa a_0$ . The experimental transition temperatures are indicated by the dotted line.

a 2D rectangular Wigner lattice is unstable. Similarly for the "y" mode, the energy change is

$$\delta V^c + \delta V^y = (C'_x q_x^2 + C'_z q_z^2) |\delta c_y(\mathbf{q})|^2 \quad (7)$$

where  $C'_x = \kappa b^2(\pi^{-1/2} + 7/4)d/(a_x a_z)$ ,  $C'_z = 0.5E_c + 3.6\kappa b^2/a_z$ . The "elastic constants"  $C$  for the x mode are much smaller than that for the y mode. When the pinning energy are included, the energy we get is of the same form as that in the study of the roughening transition.

For a single unpaired grain boundary, the elastic energy is of the **first** power in  $q$ . The cost of the long range fluctuation is higher and these fluctuations are suppressed. As a result, an unpaired grain boundary does not roughen. We next estimate the roughening temperatures of our system.

From the study of the roughening transition we find that, as the pinning Peierls potential approaches zero, the roughening transition is given by

$$kT_c = 2a_z C/a_x \quad (8)$$

where  $C$  is the geometric mean of the elastic constants. For example, for the x mode  $C = (C_x C_y)^{1/2}$ . The crystal symmetry of  $^4\text{He}$  is HCP. In the present calculation, we have approximated it by an elastically isotropic system. Our goal is not to make an accurate prediction of the temperature but to find out if the physics discussed in this paper is of

relevance to the experimental system. We have used the following estimates of the Lamé constants[11]  $\mu = 0.72K/A^3$ .  $\lambda = 1.7K/A^3$ . From these, we find  $\kappa b^2 = 3.7 \times 10^2 K \text{\AA}$ .

We have used[12]  $\theta = 0.2$  degree. The linear spacing between the dislocations is thus  $a_x = a_0/\theta = 1029 \text{\AA}$ . From an estimate of the dislocation density of  $6 \times 10^9/cm^2 = 1/(a_x d)$ , we estimate a mean spacing between the boundaries as  $d = 10^4 \text{\AA}/6$ . The core energy is not known. In two dimensional melting, it is suggested[13] that when  $E_c > E_{c0} = 0.056\kappa a_0$ , the transition becomes first order. This provides a sense of scale for  $E_c$ . In fig. 2 we show the roughening temperature for the x and the y modes (dashed and solid lines respectively) as a function of  $E_c$ . When  $E_c$  is around  $E_{c0}$ , our estimate of  $T_R$  for the x mode is of the same order of magnitude as the experimental transition temperatures of Day and coworkers[3].  $T_R$  for the y mode is of the same order as that of Burns and coworkers[2] and not a strong function of  $E_c$ . The experimental results are indicated by the dotted lines. We next examine the physical implications of the transitions.

We first show that only the upper y transition is connected with the large scale orientation fluctuation of the transition. Our calculations is connected with the fluctuation of the position of the dislocations. We first relate these to the fluctuation of the atomic positions. The displacement  $\mathbf{u}$  of the crystal at  $\mathbf{r}$  caused by a dislocation located at position  $\mathbf{c}$  can be written in Fourier transform as  $\mathbf{u}(\mathbf{r} - \mathbf{c}) = \int d\mathbf{k} \exp[i\mathbf{k} \cdot (\mathbf{r} - \mathbf{c})] \mathbf{u}_k$  where

$$\mathbf{u}(k) = b[\mathbf{e}_y/k^2 - 0.5(1 - 2\sigma)/(1 - \sigma)\mathbf{k}k_y/k^4.]$$

$\sigma$  is the Poisson ratio. The change in the displacement as the dislocations on the two boundaries are moved from  $\mathbf{c}$  to  $\mathbf{c} + \delta\mathbf{c}$  contains contributions from both grain boundaries and is equal to  $\delta\mathbf{u} = \delta\mathbf{u}_1(\mathbf{r}) - \delta\mathbf{u}_2(\mathbf{r})$ ;  $\delta\mathbf{u}_1(\mathbf{r}) = \sum_j [\mathbf{u}(\mathbf{r} - \mathbf{c}_j - \delta\mathbf{c}_j) - \mathbf{u}(\mathbf{r} - \mathbf{c}_j)]$ ,  $\delta\mathbf{u}_2(\mathbf{r}) = \sum_j [\mathbf{u}(\mathbf{r} - \mathbf{c}_j - \mathbf{e}_y d - \delta\mathbf{c}_j) - \mathbf{u}(\mathbf{r} - \mathbf{c}_j - \mathbf{e}_y d)]$ .  $\mathbf{c}_j$  is in the xy plane.

Let us look at the angular fluctuation. We have  $\delta\theta = (-\partial_x \delta u_y + \partial_y \delta u_x)/2$ . For the "parallel" wave vector  $k_p$  in the xz plan, we are interested in  $\int dy < (\delta\theta)_{k_p} (\delta\theta)_{-k_p} > = \int dk_y < |\delta\theta_{k_p, k_y}|^2 >$ . It is straightforward to show that  $\delta\mathbf{u}(k) = \mathbf{u}(k)[g(k) - g'(k)]$  where  $g - g' \approx k_y d \sum_G \mathbf{k} \cdot \delta\mathbf{c}_{\mathbf{k}_p}$ . We finally obtain, with a = x, y,

$$\int dy < (\delta\theta_{k_p})^2 > = \sum_{a, G} F_a(k) < |\delta c_a(-k_p - G) \delta c_a(k_p + G)| >. \quad (9)$$

$F_a(k) = \int dk_y [-k_x u_y(k) + k_y u_x(k)]^2 (k_y d)^2 k_a^2$ ,  $G$  is a reciprocal lattice vector. Thus the angular fluctuation of a grain can be related to fluctuation of the position of the GB. Substituting

in the expression for  $\mathbf{u}(k)$ , we get

$$F_a = b^2 k_x^2 \int dk_y (k_y d)^2 k_a^2 / k^4.$$

Thus, for  $k_z = 0$ ,

$$\int dy < (\delta\theta_{k_p})^2 > \approx k_x^3 < |\delta c_x(-k_x) \delta c_x(k_x)| > \quad (10)$$

for the x mode

$$\int dy < (\delta\theta_{k_p})^2 > \approx k_x^2 \pi < |\delta c_y(-k_x) \delta c_y(k_x)| > / a \quad (11)$$

for the y mode.

From previous calculation of the roughening transition[10], in a purely relaxational model,

$$< |\delta c(-k_x) \delta c(k_x)| > \approx 1/[A(k_x^2 + \xi^{-2})].$$

As the transition is approached, the "elastic" coefficient  $A$  and the relaxational rate are expected to exhibit square root cusps; with  $\xi \approx \exp[c/(T_R - T)^{1/2}]$  for  $T < T_R$ .

In the roughened phase with  $\xi^{-1} = 0$ ,  $< |\delta c(-k_x) \delta c(k_x)| > = 1/(Ak_x^2)$ . As  $k_x$  approaches zero,  $\int dy < (\delta\theta_{k_p})^2 >$  becomes finite above the higher temperature roughened phase but remains zero between the two roughening transitions. This is consistent with our interpretation that the higher roughening transition for the y mode corresponds to the observation from recent X-ray measurements[2] Above the x mode roughening transition, because of the change in mobility of the dislocation, a change in the elastic coefficients results; similar to that observed experimentally[3].

Experimentally the addition of  $^3\text{He}$  in ppm to ppb concentration changes the temperature dependence of the elastic modulus anomaly. The core energy can be changed. As we see in fig. 2,  $T_R$  can be a sensitive function of  $E_c$ . The impurities can also change the effective magnitude of the pinning potential as follows. We represent the pinning potential between  $^3\text{He}$  atoms at  $\mathbf{s}_a$  and the dislocation line elements by  $V^i = \sum_a W(\mathbf{c}_j - \mathbf{s}_a)$ . This appears as a factor  $[\exp(-V^i/kT)]_{av}$  in the partition function where the square bracket means an impurity averaging over the positions  $\mathbf{s}_a$ . If we approximate this factor by an cumulant expansion, we get to lowest order a term of the form  $\exp([V^i]_{av}/kT)$ . Because the  $^3\text{He}$  appears as a substitutional impurity,  $[V^i]_{av}$  has the same periodicity as the lattice and just modifies the effective strength of the Peierl's potential  $U_{x,y}$ . The next order in the cumulant expansion provides for an effective short range attraction between the dislocation lines, which

can modify the elastic constants  $C$  and hence  $T_R$ . This short range interaction is probably weaker than the long range elastic interaction in eq. (1).

The main difference between the current system and the pinning of flux line lattices (FLL) in superconductors and incommensurate charge density wave (CDW) systems is the presence of the commensurate pinning potential in the current system so that the "ground state" is simple. For the CDW and the FLL system, the focus is on the nature of the ground state as a result of the competition between the random pinning potential and the elastic energy cost to distort the lattice. In those systems, the interest is usually on three dimensional objects whereas the current system is two dimensional.

In conclusion we found that grain boundaries can roughen; there are two transitions. Estimate of their physical properties and their transition temperatures are consistent with recent experimental observations. We do not completely understand what is the connection between grain boundary roughening and supersolid behavior. We can think of many scenarios but further work is necessary to clarify if any of them is valid.

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